

A Factor Model to Explain the Hydrochemistry and Causes of Fluoride Enrichment in Groundwater of Bhairmgarh Block in Bijapur District, Chhattisgarh, India

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Abstract— High fluoride (>1.5 mg/l) in groundwater has been reported from many States of India. To understand the chemical behaviour of fluoride (F^-) in groundwater water in terms of local hydrogeological setting, aquifer level, climatic conditions and agricultural practices. In the study, a factor model was developed to determine the major hydrochemical processes that control the variations and dissolution of fluoride-bearing minerals in groundwater of Bhairmgarh block of Biapur district in Chhattisgarh. A total of 36 groundwater were collected and of F^- chemically analyzed. The results of chemical analyses (pre-monsoon 2012) show F^- abundance in the range of 0.14 to 4.7 mg/l with 25% of the samples in excess of permissible limit of 1.5 mg/l. Present study finds that four major processes control the hydrochemistry of groundwater resources in the study area: dissolution of soluble salts in the aquifer, high pH and Na^+ concentration, maximum retention time and dissolution of bicarbonate minerals in the aquifers. Two major groundwater types have been distinguished in this study: fresh Na-Ca- HCO_3 groundwater type and Na-Ca-Cl- HCO_3 groundwater type. Presence of F^- bearing minerals in the host rock, the chemical properties like decomposition, dissociation and dissolution and their interaction with water is considered to be the main cause for F^- in groundwater. The suggested remedial measures to reduce F^- pollution in groundwater include dilution by blending, artificial recharge, rainwater utilization, well-organized agricultural practices and well construction.

Index Terms— Alkalinity, Dissolution, Factor Analysis, Fluoride, Fluorosis, Groundwater

I. INTRODUCTION

The occurrence of F^- in groundwater is mainly due to geogenic factor of the groundwater and depends on the geological setting and aquifer condition of an area [1]. The F^- concentration in groundwater is originate from the dissolution of F^- bearing minerals in the bedrock and thus bedrock mineralogy plays a primary factor for the variations in F^- concentration in groundwater [2]. Including geological factor, few chemical factors also responsible for high F^- concentration in groundwater. The chemical factors such as

pH, concentration of Ca^{2+} , HCO_3^- , Na^+ ions in water. The F^- concentration > 1.5 mg/l in groundwater may persist for very long time, enters into body and cause adverse impact on human health [3]. Dental and Skeletal fluorosis are the irreversible and incurable disease, which happened due to intake of high F^- drinking water for long duration in both animal and human being [4-7].

Due to complexity of the chemical evolution of groundwater and some times substantially large amount of base information available would not provide justified explanation. The application of multivariate statistical methods helps to finds the geo-environmental problems at various scales. Although statistical associations do not establish cause-and-effect relationships. It provides relations of geo-chemical factors, which helpful to determine the geological setting and aquifer condition of an area. The uses of these advanced statistical models in recent times in the geosciences has been diverse and cover wide ranges from the resolution of simple geo-environmental problems to the determination of groundwater composition [8], the identification of factors that influence hydro-chemical changes [9] and the characterizations of hydrochemical variations among aquifer [10-11]. Factor analysis (FA), as stated by Harman (1976) 'does give a simple interpretation of a given body of data and thus affords a fundamental description of the particular variables analyzed'. FA has several advantages over classical graphical approaches in that: (1) Neutral chemical species (e.g., SiO_2) and nonchemical data (e.g., temperature, depth) can be included in the interpretation (2) variations in ions in small concentrations (e.g. Br^- , F^- , I^-) are not masked by chemically similar ions in greater concentrations (e.g., Cl^-): and (3) Secondary mixing trends are emphasized [12].

Briefly explained, the FA proceeds as follows: the correlation matrix, i.e., the array of correlation coefficients for all possible pairs of variables, is calculated. Then, the matrix is diagonalized and its principal components (eigen vectors) are obtained. The so-called factor 1 will be related to the largest eigenvalue and able to explain the greatest amount of variance in the data set. The factor 2 explains the greatest of the remaining variance, and so forth. In water quality studies [13]. FA is used in a similar way as Piper (1958) trilinear diagrams, with an added advantage that the researcher is able to include other parameters which are not usually used in conventional graphical treatment of hydrochemical data such as the Piper diagrams [14].

1.1. Study Area:

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Bhairmgarh Block comes under Bijapur district of Chhattisgarh. It located in the southern part of Chhattisgarh and lying between 80°27 E-80°30 E and 18°40 N- 19°25N. It has a population of 8.65 lakhs (as per 2001 census) in 4125 sq.Km. area as shown in Fig.1.

Bhairmagrh block falls in the semi-humid tract of the southern part of Chhattisgarh and is geologically occupied by Sabari and Pakhal group rocks (Grey granites, Charnokites, pegmatites, gneisses associated rocks etc) where groundwater occurs under unconfined condition [15]

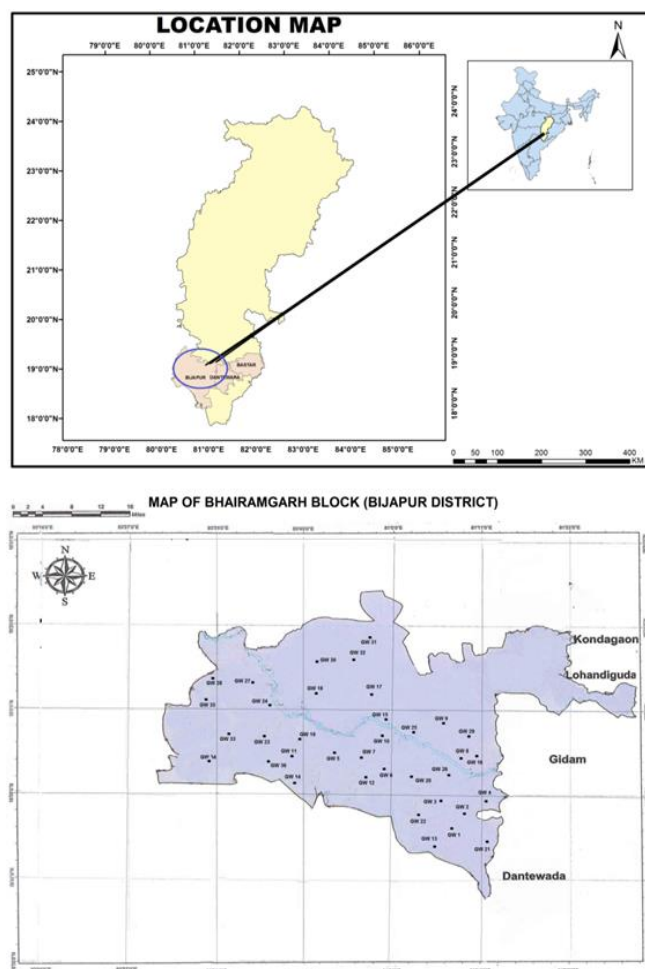


Fig.1 Map and Sampling Point of the study area

Table1: Showing the sampling point with coordinate details

Sample ID	Village Name	Latitude (East)	Longitude (North)
GW 1	Kurmer	81°4'32	18°49'12
GW 2	Tolhar	81°6'15	18°57'24
GW 3	Keskutul	81°3'11	18°58'6
GW 4	Bodali	81°11'45	18°58'25
GW 5	Potenar	80°52'3	19°1'22
GW 6	Katapal	80°58'10	19°2'35
GW 7	Matwara	80°57'18	19°4'27
GW 8	Ghotpal	81°6'29	19°6'33
GW 9	Kiskal	81°4'33	19°7'35
GW 10	Berser	80°58'26	19°1'46
GW 11	Addawalli	80°45'28	19°5'34

GW 12	Jangla	81°54'14	18°59'3
GW 13	Hawaka	81°2'36	18°49'11
GW 14	Kompalli	80°47'31	19°00'29
GW 15	Gudra	80°58'19	19°8'24
GW 16	Markapal	81°6'46	19°3'18
GW 17	Dinalur	80°58'38	19°11'5
GW 18	Bori	80°50'24	19°10'18
GW19	Jegur	80°52'32	19°3'23
GW 20	Bondapal	81°2'14	19°1'21
GW 21	Cherli	81°10'4	18°49'9
GW 22	Torinar	81°4'29	18°56'5
GW 23	Bhattapalli	80°42'11	19°5'45
GW 24	Musali	80°45'6	19°10'1
GW 25	Angwara	81°00'14	19°7'38
GW 26	Bhatwara	81°2'33	19°2'22
GW 27	Kodepalli	80°41'41	19°13'26
GW 28	Rengawaya	80°36'46	19°12'45
GW 29	Takilor	81°9'52	19°8'29
GW 30	Korangul	80°50'22	19°6'20
GW 31	Jatai	80°57'9	19°7'36
GW 32	Dompara	80°51'46	19°15'5
GW 33	Alwara	80°39'37	19°7'13
GW 34	Kupral	80°37'8	19°5'17
GW 35	Doke	80°35'19	19°9'34
GW 36	Ponarwaya	80°46'34	19°1'30

II. EXPERIMENTAL MATERIALS

2.1.Materials and Methods

Water samples were collected in pre-cleaned 1L sampling polyethylene container from 36 groundwater sources (Manully handpumps). Before taking final sample, the water was left to run from sampling sources for 10 min to pump out the volume of water standing in casing. The phyco-chemical parameters such as, temperature, pH and conductivity were measured at the time of sampling [16]. The pH was measured by pocket pH meter model Eutech, Model-pH tester 34 and conductivity by conductivity meter model Eutech, Model 824 respectively. All the chemical used were of AR grade (Fischer, Scientific, India and Merck, Germany) and Milli-Q water (Milipore Corp. With electrical resistivity of 18.2 MΩ cm) was used for preparations of solutions and dilution purposes. Table 2 provide the procedure and instrument used for the analysis of groundwater samples [17].

Table 2:Details of Physiochemical parameters with detail of analyzed method.

Parameters	Instrument/Method	Method for Analysis
Temp.	Thermometer	

pH	Eutech, Model-pH tester 34 (Pocket pH meter)/Elico Model 1012	Potentiometric
EC,	Eutech, Model 824/Model No-304 Systronics	Potentiometric
TH	EDTA method	Titrimetric
Ca ²⁺	EDTA method	Titrimetric
Mg ²⁺	EDTA method	Titrimetric
TA		Titrimetric
HCO ₃ ⁻		Titrimetric
Na ⁺	Elico CL-361	Flame Photometer
K ⁺	Elico CL-361	Flame Photometer
TDS		Gravimetric
NO ₃ ⁻	Nova 60 Spectroquant® photometer	Photometer
Cl ⁻	Silver nitrate method	Titrimetric
F ⁻	Nova 60 Spectroquant® photometer and Orion Ion Selective electrode	Photometer & Ion selective electrode
SO ₄ ²⁻	Nova 60 Spectroquant® photometer	Photometer

Note:- All parameters are in mg/l except Electrical conductivity (EC) is $\mu\text{S}/\text{cm}$ TH= Total Hardness, TA= Total Alkalinity

Statistical characterization of data and correlation matrix (Pearson's correlation) was analyzed using Graphpad Prism (Version 5.03). Factor analysis was carried out using XLSTAT 2015 (Version 17.4.01.22222). Factor analysis using Principal components analysis (PCA) was applied only those Eigen values are greater than one.

III. RESULTS AND DISCUSSION

3.1 General hydrochemistry and F⁻ enrichment

Statistical summaries of the physico-chemical parameters used in this study for the 36 samples presented in Table 2. The parameters are very variable in terms of their concentrations in the entire study area. The electrical conductivity (EC) ranges from a low value of 368 $\mu\text{S}/\text{cm}$ to 1106 $\mu\text{S}/\text{cm}$ which represent fresh and brackish groundwater types as they respectively represent total dissolved solid (TDS) concentration of 290 mg/l to 632 mg/l. the variability in the EC value is indicated by the high standard deviation (Table - 2).

Among anions HCO_3^- (range 146-285 mg/l, mean \pm SD, 205.3 \pm 42.6) is present in higher concentration, Cl^- (range 26-157 mg/l, mean \pm SD, 79.2 \pm 35.4), SO_4^{2-} (range 18-133 mg/l, mean \pm SD, 69.4 \pm 33.7), TA (range 120-234 mg/l, mean \pm SD, 168.38 \pm 34.94) and F^- (range 0.14-4.7 mg/l, mean \pm SD, 1.44 \pm 0.89) are present in low to moderate concentration. It might be due to presence of some saline beds at some locations. High F^- levels in groundwater are one of the major concerns in some areas within the study area. The average F^- concentration in the area 1.44 mg/l, which is within permissible limit <1.5 mg/l [3]. From analysis it was found that western part of the study area reported high range of F^- concentration, studies suggest that the high F^- is

attributed to the dissolution of fluoride in the aquifer [18,1]. Hydrochemical analysis of the groundwater indicate the presence of fluorite in appreciable quantities to cause such high F^- levels in the groundwater. In the Bhairmgarh Block and its surrounding areas, the sources of high F^- in groundwater have not yet been confined. On the basis of extensive analysis for F^- it was found that 26.6% sample out of 30 samples exceeds maximum permissible limit of WHO, 2006 (>1.5 mg/l). Groundwater samples of 9 villages contain F^- concentration >1.5 mg/l. The villages where high F^- reported, namely Katapal (3.2 mg/l), Berser (3.0 mg/l), Addawalli (2.2 mg/l), Jangla (4.7 mg/l), Markapal (2.3 mg/l), Bhattapalli (1.8 mg/l), Kodepalli (2.6 mg/l), Alwara (2.0 mg/l) and Kupral (1.9 mg/l) respectively.

Among cations, Ca^{2+} (range 26-73 mg/l, mean \pm SD, 44.5 \pm 10.8), Mg^{2+} (range 16-53 mg/l, mean \pm SD, 32.8 \pm 9.7), Na^+ (range 1.5-114 mg/l, mean \pm SD, 46.80 \pm 38.73) is present in higher concentration whereas K^+ (range 0.6-55.9 mg/l, mean \pm SD, 11.74 \pm 14.50) is present in moderate concentration.

Table 3: Statistical summaries of the major parameters used for the study of the hydrochemistry.

Variable	Min.	Max.	Mean	Std. Dev.
pH	6.74	8.32	7.61	0.36
EC $\mu\text{S}/\text{cm}$	368.00	1106.07	769.60	192.29
TDS (mg/l)	290.06	632.04	442.35	103.50
Mg^{2+} (mg/l)	16	53	32.80	9.77
Ca^{2+} (mg/l)	26	73	44.57	10.80
Na^+ (mg/l)	1.5	114	46.80	38.73
K^+ (mg/l)	0.60	55.9	11.74	14.50
Cl^- (mg/l)	26	157	79.2	35.4
HCO_3^- (mg/l)	146.0	285.56	205.3	42.6
SO_4^{2-} (mg/l)	18	133	69.4	33.7
F^- (mg/l)	0.14	4.7	1.44	0.89
NO_3^- (mg/l)	11.20	76.20	32.64	18.08
TA (mg/l)	120	234	168.38	34.94
TH (mg/l)	172.90	387.30	245.90	55.73

Boxplot were used to represent temporal concentration of the major ions (Figure 2). The upper and lower quartiles of the data define the top and bottom of rectangle box. The line inside the box represents the median value and the size of the box represents the spread of the central value [19]. The concentration values of ions like Na^+ , Mg^{2+} , Ca^{2+} and HCO_3^- was found to increase due to the effective leaching from rock

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matrix [20]. HCO_3^- is mainly formed due to the action of atmospheric CO_2 and CO_2 released from organic decomposition [21]. Concentration of SO_4^{2-} indicates lithogenic sources of sulphate noted in the study area [20]. NO_3^- in groundwater derived from organic industrial effluents, fertilizers or nitro-fixing bacteria, leaching of animal dung sewage and septic tanks through soil and water matrix to groundwater. In general, increase of nitrate in groundwater may be an indicator of biological pollution [22-23]. The large variations in mean, median and standard deviation values of ions suggest that the water chemistry in the study region is heterogeneous and influenced by complex hydro chemical process and complex contaminates sources [24]. From boxplot it is clear that among cation Na^+ become dominant anions HCO_3^- . Thus, the order of their concentration in groundwater is as given below.

Cations- $\text{Na}^+ > \text{Ca}^{2+} > \text{Mg}^{2+} > \text{K}^+$

Anions- $\text{HCO}_3^- > \text{Cl}^- > \text{SO}_4^{2-} > \text{NO}_3^- > \text{F}^-$

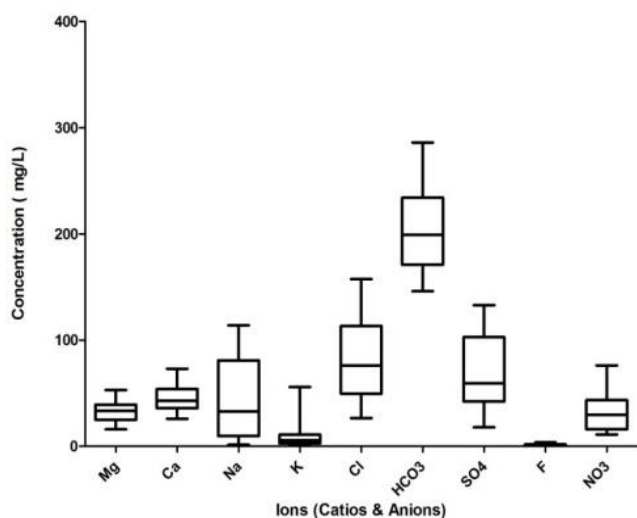
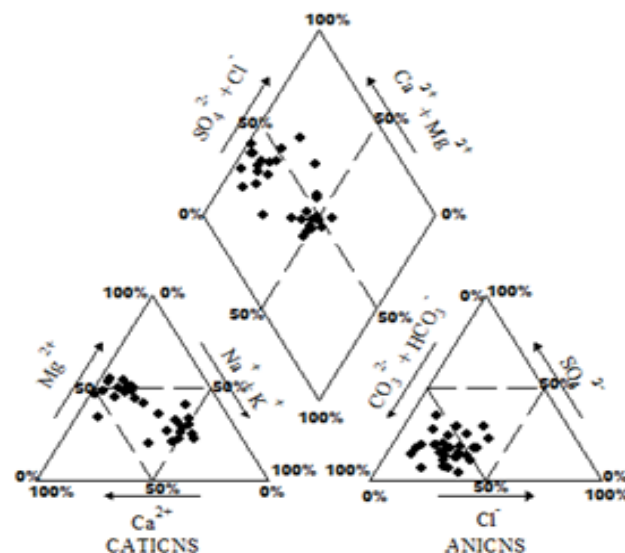
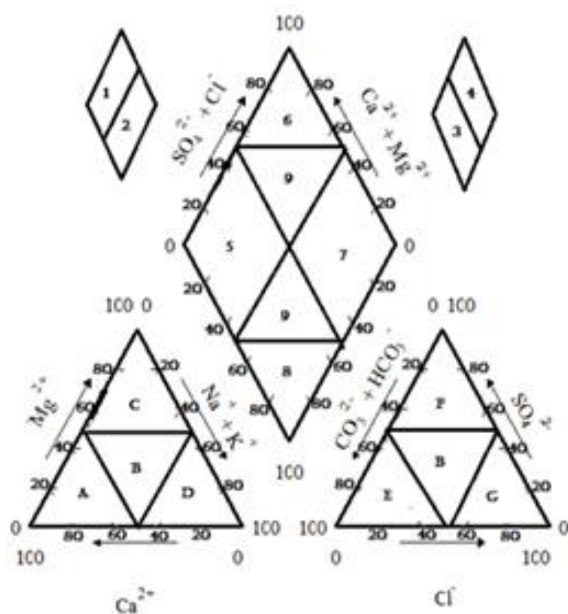


Fig. 2 Boxplot for the cations and anions of the study area.



Legend:

- A. Calcium type
- B. No dominant type
- C. Magnesium type
- D. Sodium and potassium type
- E. Bicarbonate type
- F. Sulphate type
- G. Chloride type

Fig. 3. Piper trilinear plot of the samples of the study area with legend .

Table 3: Characterization of groundwater of Bhairmgarh Block on the basis of Piper-tri-linear diagram.

Sub division of the diamond	Characteristics of corresponding subdivisions of diamond-shaped fields	Percentage (%) of samples in the category
		Pre-Monsoon Season
1	Alkaline earth (Ca +Mg) Exceed alkalies (Na+K)	90
2	Alkalies exceeds alkaline earths	10
3	Weak acids ($\text{CO}_3 + \text{HCO}_3$) exceed Strong acids ($\text{SO}_4 + \text{Cl}$)	80
4	Strong acids exceeds weak acids	20
5	Magnesium bicarbonate type	65.5
6	Calcium-chloride type	0
7	Sodium-chloride	4.5
8	Sodium-bicarbonate	0
9	Mixed type (No cation-anion exceed 50%)	30

The ionic concentration of major cations and anions found in groundwater of the study area are plotted in Piper's trilinear diagram (Figure 3). Among the total of 30 groundwater samples, cations are clustered within the area of Ca + Mg covers the area of 90% and Na+K, while anions fall within the area of 80% $\text{HCO}_3^- + \text{CO}_3^{2-}$ and 20% $\text{Cl}^- + \text{SO}_4^{2-}$. According to (Apambire 1997) groundwater with high F^- concentration is generally of HCO_3^- -Na type, particularly with poor Ca^{2+} . Several authors have reported that in water with high F^- concentration, the amount of F^- is proportional to the

HCO_3^{2-} concentration and pH [25,1]. It was observed in the present study that the elevated F^- concentration (>1.5 mg/l) is associated with mixed water group where Na^+ concentration is relatively higher than other cations. Such mixed group types are Na-Ca-Mg-Cl- HCO_3 , Mg-Na- CO_3 - HCO_3 -Cl, Mg-Na- HCO_3 - SO_4 and Mg-Na- HCO_3

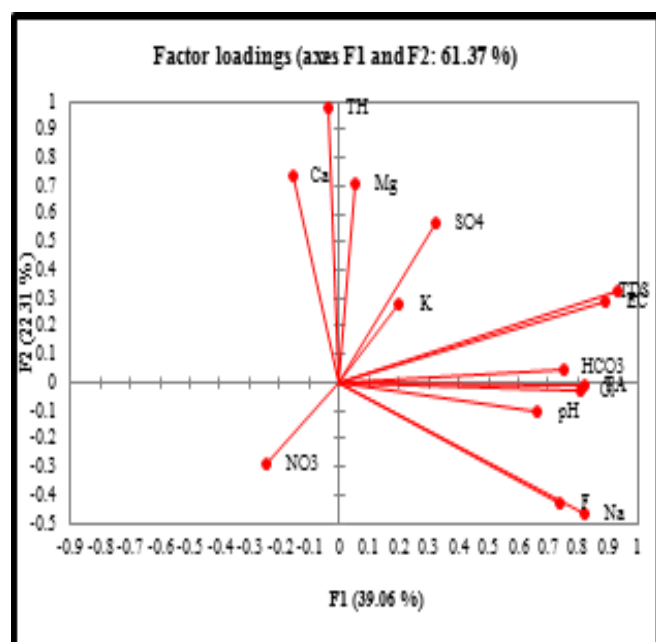


Fig. 4 Factor loadings (axes F1 and F2: 61.37%) of different hydrochemical parameters (GW-36)

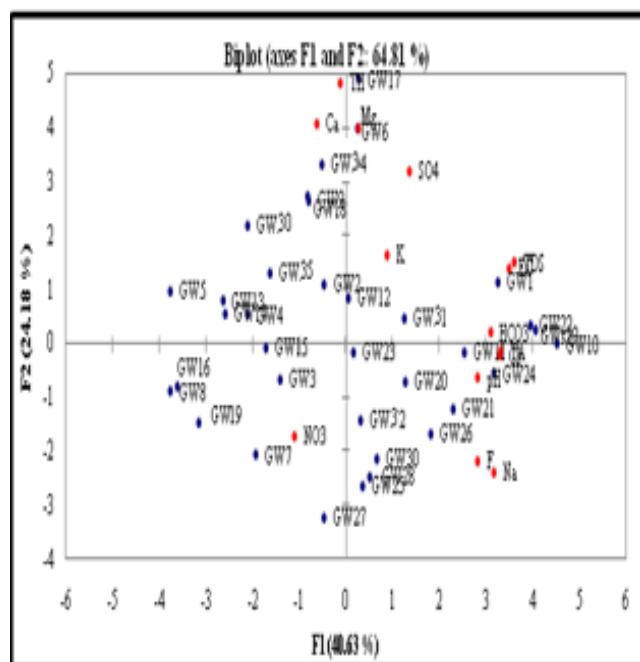


Fig. 5 Principle component analysis for water quality parameters (Factor observation axes F1 and F2: 64.81%).

In order to further investigate and identify contributing sources on the basis of chemical signatures, factor analysis (FA) of the data was done. The analysis yielded four dominant factors and was expressed by data matrix variance of 61.37%. Factor loading is classified as 'strong', 'moderate' and 'weak'

corresponding to absolute loading values (positive or negative) of >0.70 , $0.70-0.45$ and $0.45-0.25$ respectively. Component loading of principle components is shown in figure 3.

Factors 1 explains 40.63% of total variance showing loadings of Na^+ , SO_4^{2-} , EC, TDS, pH, Cl^- , K^+ , TA, HCO_3^- and Mg^{2+} . Factors shows the strong loading of TDS, Na^+ , EC, F^- , HCO_3^- , Cl^- , pH and TA, and weak loading of K^+ and SO_4^{2-} . The combinations of these factors represents the dissolved ion load of groundwater [26]. Thus from the results it is clear that Na^+ , Mg^{2+} , F^- , HCO_3^- and Cl^- ions contributes to the ion load of groundwater [27].

Factor 2, explains 24.31% of total variance. Strong positive loading show by Ca^{2+} , Mg^{2+} and Total hardness (TH). However total hardness is due to both Ca^{2+} and Mg^{2+} ions, moderate loading of Na^+ and SO_4^{2-} and weak loading of EC, TDS, K^+ , F^- and NO_3^- .

Factor 3, explains the 9.99% of total variance. Factors shows strong loading of K^+ , moderate loading of NO_3^- and weak loading of F^- and SO_4^{2-} . Factor 4, explains of 6.93% of total variance, None of the parameters shows any significance between themselves. Factor 5, explains the 6.33% of total variance. Significant correlation between total alkalinity (TA) and HCO_3^- has been observed. Factor 6, explains of 3.83% of total variance. None of the parameters shows any significance between themselves.

Table 4: Factor loading of different chemical parameters of groundwater samples (GW-30) from the study area (Significant loadings marked in bold).

Note:- Except pH and EC all parameters are in mg/l. EC= $\mu\text{S}/\text{cm}$

Parameter	F1	F2	F3	F4	F5	F6
pH	0.719	-0.125	-0.098	0.248	-0.151	0.51
EC	0.894	0.276	-0.047	-0.076	-0.132	-0.179
TDS	0.924	0.296	-0.088	-0.087	-0.151	-0.084
Mg^{2+}	0.068	0.786	0.047	0.45	0.26	-0.148
Ca^{2+}	-0.158	0.806	0.21	-0.394	-0.08	0.154
Na^+	0.812	-0.469	-0.132	0.014	-0.19	-0.05
K^+	0.224	0.327	0.727	0.275	-0.328	0.153
Cl^-	0.842	-0.041	-0.013	0.245	-0.146	-0.326
HCO_3^-	0.791	0.039	0.122	-0.243	0.498	0.149
SO_4^{2-}	0.347	0.63	-0.32	-0.393	-0.367	0.02
F^-	0.721	-0.428	0.417	0.05	0.067	0.021
NO_3^-	-0.28	-0.338	0.683	-0.337	-0.126	-0.189
TA	0.851	-0.027	0.104	-0.236	0.4	-0.008
TH	-0.028	0.955	0.136	0.132	0.149	-0.032
Eigen value	5.689	3.385	1.4	0.971	0.887	0.537
Variability (%)	40.634	24.175	9.997	6.936	6.337	3.834
Cumulative %	40.634	64.809	74.807	81.743	88.08	91.913

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Table 5: Correlation matrix of hydrochemical parameters of groundwater samples of study area.

Variables	pH	EC	TDS	Mg ²⁺	Ca ²⁺	Na ⁺	K ⁺	Cl ⁻	HCO ₃ ⁻	SO ₄ ²⁻	F ⁻	NO ₃ ⁻	TA	TH
pH	1													
EC	0.49	1												
TDS	0.59	0.95	1											
Mg ²⁺	0	0.19	0.23	1										
Ca ²⁺	-0.24	0.08	0.1	0.36	1									
Na ⁺	0.67	0.59	0.66	-0.32	-0.54	1								
K ⁺	0.15	0.3	0.26	0.27	0.3	-0.03	1							
Cl ⁻	0.57	0.8	0.78	0.1	-0.22	0.73	0.19	1						
HCO ₃ ⁻	0.48	0.64	0.66	0.08	0.01	0.5	0.08	0.49	1					
SO ₄ ²⁻	0.18	0.54	0.62	0.3	0.5	0.12	0.05	0.16	0.18	1				
F ⁻	0.52	0.47	0.49	-0.21	-0.4	0.73	0.3	0.58	0.62	-0.14	1			
NO ₃ ⁻	-0.26	-0.35	-0.35	-0.31	-0.01	-0.1	0.13	-0.23	-0.17	-0.3	0.18	1		
TA	0.47	0.73	0.74	0.03	-0.09	0.6	0.09	0.59	0.92	0.18	0.65	-0.15	1	
TH	-0.12	0.18	0.21	0.89	0.74	-0.49	0.34	-0.03	0.06	0.46	-0.34	-0.23	-0.02	1

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To examine the relationships of F⁻ with other geochemical parameters, correlation matrix and scatter plot have been generated for derived parameters of groundwater samples. The correlation matrix (Table 5) exhibits excellent positive correlation among pH, TDS, Na⁺, K⁺, TA and Cl⁻. This is due to the fact that conductivity depends on total dissolved solids and main constituents of TDS in water are Na⁺, Cl⁻, Ca²⁺ and HCO₃⁻. Also from correlation matrix it is found that Ca²⁺, Mg²⁺, HCO₃⁻ and Cl⁻ like soluble salts are predominant in groundwater of the study area. F⁻ has negative correlation with Ca²⁺, Mg²⁺, TH and SO₄²⁻. This is in accordance with observation made earlier (previous section) that as F⁻ concentration increases, Na⁺ concentration increase and Ca²⁺ and Mg²⁺ concentration decreases [28].

IV. CONCLUSION

The hydrochemistry of Bhairmgarh block highlight that high F⁻ in the groundwater are due to geogenic factor. Including that high pH, alkalinity and bicarbonate content promote the weathering of F⁻ bearing minerals. A factor model from the hydrochemical data suggests that F⁻ enrichment is associated with positive loading of HCO₃⁻ and Na⁺ and negative loading of Ca²⁺ and Mg²⁺ might be favour the weathering of genesis and granite rocks which contain F⁻ minerals. To know hydro-chemical control in relation to aquifer character isotope studies may be taken up for aquifer mapping to delineate the F⁻ rich minerals and artificial flushing of identified aquifer to reduce F⁻ concentration.

ACKNOWLEDGMENT

The authors are thankful to the National Institute of Technology, Raipur, India for providing all necessary facilities.

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